

THE EFFECTS OF CLOUDY/CLEAR AIR MIXING AND
DROPLET pH ON SULFATE AEROSOL FORMATION
IN A COUPLED CHEMISTRY/CLIMATE GLOBAL MODEL

C. R. Molenkamp, J. E. Penner, J. J. Walton, and C. A. Atherton
Atmospheric Sciences Division
Lawrence Livermore National Laboratory
Livermore, CA USA

Sulfate aerosols affect global climate by scattering solar radiation and modifying the cloud drop size distribution, which further changes the back-scattering of solar radiation and the cloud life cycle. The negative climate forcing of aerosols counteracts the warming associated with increased greenhouse gases; however, the pattern of forcing is quite different because the distribution of sulfate aerosols is regionally inhomogeneous. Most sulfate aerosols form in the atmosphere through gas phase reactions of DMS, H_2S , and SO_2 with OH or aqueous reaction of SO_2 with H_2O_2 and O_3 in cloud drops.

We have coupled our atmospheric chemistry/transport model, GRANTOUR, with the ECHAM3 global climate model which provides several enhanced capabilities in the representation of aerosol interactions. ECHAM includes a specific representation of liquid water in large-scale clouds that allows us to represent the aqueous conversion of SO_2 to sulfate as well as to improve the parameterization of precipitation scavenging. We perform calculations of the gas-phase reactions throughout the atmosphere and calculations of the gas-phase and aqueous reactions inside clouds. To represent the mixing of clear and cloudy air we periodically combine the concentrations of the reacting species based on the large-scale cloud fraction. We will present global simulation results using different mixing periods to investigate the effects of mixing time on sulfate distributions.

The volatility of SO_2 and therefore the rate of aqueous formation of SO_4^{2-} is dependent on the pH of cloud droplets. Since our model does not currently include all the species that determine pH we specify a droplet pH. We will present global simulation results with several different assumed pH's to show the effect of droplet pH on global sulfate distributions.

Acknowledgement. This work was performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore National Laboratory under contract No. W-7405 -ENG-48.